

USEtox relevance as an impact indicator for automotive fuels. Application on diesel fuel, gasoline and hard coal electricity

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Abstract

Purpose In order to provide more sustainable fuels and address the depletion of oil as a feedstock, the automotive industry must adapt to a growing market share of alternative fuels. The environmental impacts of the automotive industry to date would suggest that these alternatives will be more environmentally friendly than petroleum-based fuels. This is nonetheless an assumption that cannot be confirmed without a systematic life cycle assessment (LCA). This article explores the feasibility of USEtox to provide information needed for automotive-fuel LCA.

Materials and methods USEtox is tested on three energy pathways: gasoline, diesel fuel and hard coal electricity. The studied emissions are mainly volatile organic compounds (VOCs) and heavy metals. USEtox being dependent on the physicochemical and toxic properties of the studied species, a speciation of all VOCs emitted was performed. Moreover, since USEtox allows a distinction between rural and urban emissions, a geographical information system was developed in order to distinguish these emissions. Finally, because crude oil comes from various countries, characterization factors have been calculated for new regional compartments.

Results and discussion Human health issues are caused by aldehydes and heavy metals while ecotoxicity is caused by polycyclic aromatic hydrocarbons, aldehydes and heavy metals. For organic compounds, a clear distinction is observed between urban and rural emissions while inorganic mechanisms are independent of this distinction. Among the three energy pathways, urban diesel is the more impacting.

Conclusions USEtox can be used for the assessment of automotive fuels, though it only addresses specific aspects of human health and ecotoxicity. The LCA practitioner must keep in mind that USEtox has to be used in conjunction with other indicators, such as ReCiPe or CML, to comprehensively cover the toxic and ecotoxic impacts of fuels. The level of analysis is dependent on the accuracy of the inventory, aldehydes and PAH playing a crucial role. Inorganic impacts are highly uncertain, contrary to organic compounds. The distinction between rural and urban emissions allows a better assessment of internal combustion engine-powered cars compared with electric and hybrid cars, which is especially useful for the automotive industry now that these technologies are clearly being developed.

Keywords Automotive fuels · Hard coal power · Heavy metals · Urban pollution · USEtox · Volatile organic compounds

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Glossary

EU27	European Union
GIS	Geographic information system
GRUMP	Global Rural/Urban Mapping Project
ICE	Internal combustion engine
LCA	Life cycle assessment
NEDC	New European driving cycle
NMVOCs	Non-methane volatile organic compounds
PAH	Polycyclic aromatic hydrocarbons

PM	Particulate matter
TTW	Tank to wheels
WTT	Well to tank
WTW	Well to wheels

1 Introduction

1.1 Context and scope

Until the end of the twentieth century, the automotive industry was almost exclusively dependent on one resource: oil. However, this situation has led to five major issues: the high dependence of the European Union (EU27) to oil (which is mainly imported from outside the EU), global warming, depletion of fossil resources (amongst them oil), urban pollution and high costs of energy. To cope with these new challenges, carmakers and the energy sector are developing new alternatives for a more sustainable individual mobility. These alternatives include for instance biofuels, synthetic fuels, hydrogen or electricity. As a way to address

the five issues described above, they should be more environmentally friendly than conventional fossil fuels. Life cycle assessment (LCA) is an appropriate tool to compare the impacts of these alternatives on the environment and to compare them to conventional fossil fuels. LCA is a normalized tool (ISO14040–14044) which sums aquatic, atmospheric and soil emissions of a system, from its cradle to its grave, and calculates various associated environmental impacts, such as anthropogenic global warming, ozone depletion, acidification, or water eutrophication. This article is focused on human health issues and aquatic ecotoxicity linked with three fuel systems used in the EU27 in 2011: gasoline, diesel fuel and electricity coming from hard coal. The functional unit is 1-km driven on a new European driving cycle (NEDC). The car maintenance is considered insignificant (Renault 2009) and, for the electric vehicle, the battery manufacturing and disassembling is not considered, due to a lack of reliable data. Therefore, the system is composed of the two following stages: the “well-to-tank” (WTT) stage, meaning the production of the fuel, and the “tank to wheels” (TTW) stage, corresponding to the vehicle driven on 1 km. The sum of WTT+TTW stages, called a “well-to-wheels” (WTW) analysis, is calculated as follows:

$$\left[(\text{fuel production impacts}(\text{MJ}^{-1}) \times \text{fuel consumption}(\text{MJ km}^{-1}))_{\text{WTT}} + (\text{exhaust pipe impacts}(\text{km}^{-1}))_{\text{TTW}} \right]_{\text{WTW}}$$

1.2 Human health and ecotoxicity impacts

Numerous methods can be found in the literature to assess toxicity and ecotoxicity in LCA. Midpoint indicators including these aspects are for instance CML 2001 (Guinée 2002), EDIP 1997 (Hauschild and Potting 2005), IMPACT 2002+ (Jolliet et al. 2003) or TRACI (Bare 2002). Yet, the results obtained with these methods are difficult to interpret because of their high uncertainties. Switching from a method to another can lead to different conclusions because toxicity and ecotoxicity impacts are caused by complex mechanisms involving pollutant dispersion and reaction of the receiving ecosystems. In addition, they can be measured with several indicators (chronic or acute toxicity, no-observed effect concentration, lethal concentration 50). To illustrate these variations, Tables 1 and 2 compare two simple systems, diesel fuel and gasoline, and show, according to different methodologies, the share of the impacts caused by the car (TTW) relative to the total impacts (WTW). Data are calculated using GaBi software¹ and Querini et al. (2011). For aquatic ecotoxicity, CML 2001

freshwater aquatic ecotoxicity potential (kg DCB_{eq}), EDIP 1997 ecotoxicity (water, chronic; m³ water) and IMPACT 2002+ aquatic ecotoxicity (midpoint indicator, kg TEG_{eq}) were retained. For human health, CML 2001 human toxicity potential (kg DCB_{eq}), EDIP 1997 human toxicity in air (m³ air) and IMPACT 2002+ carcinogens and non-carcinogens aggregated impacts (kg C₂H₃Cl_{eq}) were selected. The results illustrate the variations among the three methods.

1.3 USEtox and aim of the study

Because the aforementioned methods do not converge for the two studied fuels, Renault has always been reluctant to arbitrarily choose one. USEtox (Rosenbaum et al. 2008),

Table 1 Comparison of TTW shares of diesel fuel and gasoline WTW impacts among three ecotoxicity indicators

Ecotoxicity method	TTW/WTW diesel fuel (%)	TTW/WTW gasoline (%)
CML 2001	90	65
EDIP 1997	20	33
IMPACT 2002+	30	29

¹ PE, LBP: GaBi 4™ Software-System and Databases for Life Cycle Engineering. Stuttgart, Echterdingen 1992–2000

Table 2 Comparison of TTW shares of diesel fuel and gasoline WTW impacts among three toxicity indicators

Human health method	TTW/WTW diesel fuel (%)	TTW/WTW gasoline (%)
CML 2001	36	78
EDIP 1997	89	86
IMPACT 2002+	70	95

resulting from a consensus among these various methodologies (Hauschild et al. 2008), appears to be promising to assess the human health issues and ecotoxicity associated with alternative energy sources. Therefore, the aim of this article is to assess if USEtox can be used for such a purpose, by conducting the LCA of diesel fuel, gasoline and hard coal electricity. This article also studies the influence of separate urban and rural emissions. Finally, the effect of modifying the environmental data of the continental scale to take into account oil and hard coal importations is also evaluated.

2 Materials and methods

2.1 Well-to-tank stage

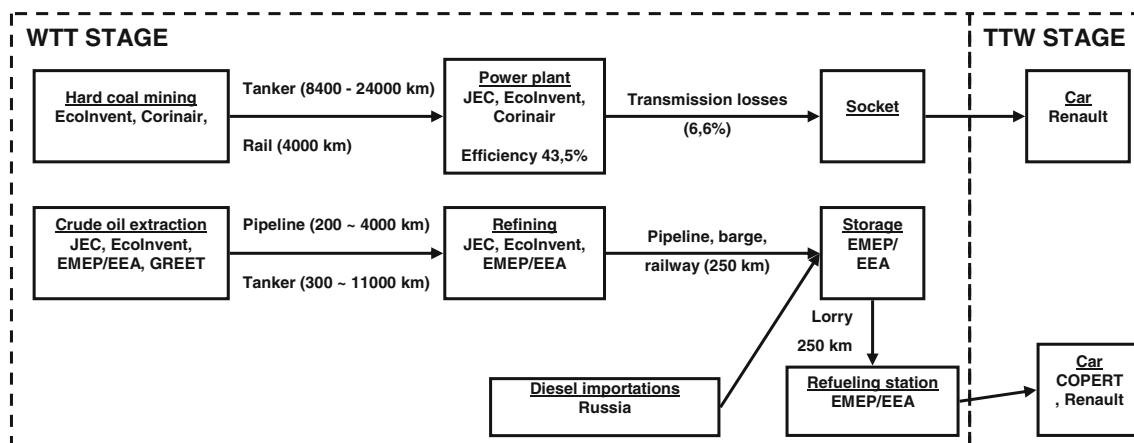
For internal combustion engine (ICE)-powered cars (diesel and gasoline), WTT pathways are mainly calculated using Edwards et al. (2009), the Ecoinvent database (Frischknet 2008), the European life cycle database (Ecobilan 2008) and the EMEP/EEA emission guidebook (European Environment Agency 2009), as described in Querini et al. (2011). The same approach is used to describe the production of hard coal electricity. Only hard coal is considered in this article, lignite (mainly produced in Germany) and anthracite being neglected. As shown on Fig. 1, the hard coal electricity pathway can be separated

in six steps: *hard coal mining* (according to Eurostat values (2008), hard coal in EU27 comes from Eastern Europe (32%), Western Europe (15%), Russia (15%), Australia (11%), South Africa (10%), USA (9%) and Latin America (8%)), *hard coal transport to EU27* (mainly by rail in Europe and from Russia and by tanker for transcontinental transport), *electricity generation* (using thermal energy from hard coal combustion), *electricity transport* (with associated losses), *socket distribution* and *car use*. Figure 1 also shows the diesel and gasoline pathways, taken from Querini et al. (2011).

Two main groups of pollutants addressed by USEtox (Huijbregts et al. 2010a) are emitted in the studied systems: non-methane volatile organic compounds (NMVOCs) and heavy metals (inorganics). Usually, NMVOC speciation is not considered in LCA databases (such as in the European life cycle database) and they are all grouped in a single emission. Even so, since NMVOC toxicity and ecotoxicity depend on the considered species, it is necessary to have an assessment of these latter. In this article, Passant (2002) is used to determine the NMVOC speciation for more than 95% of all NMVOCs emitted during the WTT stage. Heavy metals in air are calculated by applying the EMEP/EEA guidebook emission factors, and Ecoinvent is the reference for aquatic emissions. For heavy metals, a Monte Carlo analysis is conducted, using the confidence intervals given by the EMEP/EEA guidebook.

2.2 Tank-to-wheels stage

In order to calculate TTW emissions and fuel consumption, two approaches can be found in the literature: measurements on actual vehicles or defining a vehicle which is representative of a given fleet. In this article, two ICE vehicles which represent the average fleet in circulation in the EU27 in 2011 are retained, respectively for diesel and gasoline. These two vehicles are calculated using the

**Fig. 1** WTT pathways of studied fuels

NEDC cycle and Euro standards, according to Querini et al. (2011). Euro standards define pollutant thresholds at the exhaust pipe of the cars sold in the EU27 and the current version is Euro5, each new version applying higher reductions of emissions. In 2011, the average fleet is approximately composed of: Euro2 (24%), Euro3 (34%), Euro4 (28%) and Euro5 (14%), leading to the values of NMVOC emissions as shown in Table 3. For battery-powered vehicles, only a few vehicles are circulating in 2011. Therefore, a Renault electric Fluence model is chosen as the average vehicle sold in 2011 and, as a consequence, the results are only applicable for this Renault model. Obviously, NMVOCs for this vehicle are set to zero since electric vehicles do not emit any pollutant during their use phase.

For every system, toxicity and ecotoxicity impacts come from four types of pollutants: NMVOCs, heavy metals, particulate matter (PM) and nitrogen dioxide (NO₂). SO₂ used to be an issue for car exhaust emissions (by creating acidification and secondary PMs) but, since the fuel sulphur content is now limited to 10 ppm in the EU27 (Directive 2009/30/EC), it can be neglected in our study. PM exact composition is not known and therefore their toxicity is not assessed by USEtox, though there were attempts to do so (PM toxicity is both linked with their nature and size). Irritation of the pulmonary airway caused by NO₂ and ozone is also not considered by USEtox. Thus, USEtox only addresses one of the different aspects of the toxicity issues associated with road traffic and this article is, when referring to human health, only considering these impacts.

Atmospheric heavy metals mainly come from the combustion of fossil fuels because they contain small amounts of mercury, cadmium, lead and other metals which are released to water and soil when leakages occur and to air when they burn (released as metal oxides and/or adsorbed on PM). The emitted amounts can be calculated using the heavy metal contents of fuels. According to the EMEP/EEA guidebook, heavy metals emitted at the exhaust pipes of the cars are as follows (per kg of fuel burnt): cadmium (0.01 mg kg⁻¹), copper (1.7 mg kg⁻¹), chromium (0.05 mg kg⁻¹), nickel (0.07 mg kg⁻¹), selenium (0.01 mg kg⁻¹) and zinc (1 mg kg⁻¹). All these metals are included in USEtox.

NMVOC speciation comes from the COPERT project values (Gkatzoflias et al. 2007) which can be considered in

EU27 as the reference for unregulated emissions (i.e. VOC speciation and differentiation between NO and NO₂) of exhaust pipes. The values in COPERT are based on the Artemis European project (Andre 2004), which analysed a large sample of European vehicles in order to assess the regulated and unregulated pollutants on various customer cycles. Other reference study can be found in the literature (Schauer et al. 2002, Duffy et al. 1999, Liu et al. 2008, Schmitz et al. 2000) and have been reviewed by Cai and Xie (2009). Using these values allows to calculate a standard deviation (in brackets in Table 4) associated with every NMVOC species but polycyclic aromatic hydrocarbons (PAH), which are only addressed by COPERT. These standard deviations can afterwards be used to run a Monte Carlo analysis whose results are shown in Table 4 (in brackets). Table 4 also presents the values from COPERT, which offers the most comprehensive description of car exhaust by considering 91 species in the following groups.

2.3 Missing NMVOCs

Some NMVOC species emitted by the three systems are not addressed by USEtox. These substances are emitted in such quantities so that neglecting them does not lead to any variation in the results. Yet, in order to be as accurate as possible, we calculated new interim factors for them. These values were computed according to the methodology given by Huijbregts et al. (2010a). Missing substances are: benzaldehyde, methacrolein, propionaldehyde, benzofluoranthene, perylene, triphenylene, 3,6-dimethylphenanthrene, anthanthrene, coronene, propadiene, pentene, butane, terpenes, methyl borate and acenaphthene. Some of these substances have very low toxicity potentials (for instance butane or terpene) and for that reason the calculated characterization factor values are insignificant. For example, benzaldehyde's characteristics are: MW=106.13 g mol⁻¹, KOW25=30.2, Koc=11.9 L kg⁻¹, KH25C=2.71 Pa m⁻³ mol⁻¹, Pvp25=169 Pa, Sol25=6,950 mg L⁻¹, kdegA=3.40×10⁻⁶ s⁻¹, kdegW=3.40×10⁻⁶ s⁻¹, kdegSd=3.78×10⁻⁷ s⁻¹, kdegSI=1.70×10⁻⁶ s⁻¹, avlogEC50=0.157 mg L⁻¹, ED50inh (non-cancer)=46.4 kg lifetime⁻¹, ED50ing (non-cancer)=284 kg lifetime⁻¹, ED50inh (cancer)=365 kg lifetime⁻¹, ED50ing (cancer)=365 kg lifetime⁻¹, BAFfish=3.57 L kg fish⁻¹. Examples of the obtained characterization factors are, for human health: emissions to urban air (total)=3.2×10⁻⁷ cases kg⁻¹ or, for aquatic ecotoxicity, emission to continental air=18.9 PAF m⁻³ day⁻¹ kg⁻¹.

2.4 Continental differentiation

USEtox considers three compartments: urban, continental and global. The global compartment represents the whole world and that is why it is not modified. However, the

Table 3 Fuel consumption and NMVOC emissions of the three cars considered

Vehicle (2011)	Gasoline— average fleet	Diesel— average fleet	Electric vehicle
Consumption (MJ/km)	2.57	2.53	0.47
NMVOCs (mg/km)	228	77	0

Table 4 Mass composition of NMVOCs from gasoline and diesel exhaust gases (from COPERT values)

Species	Gasoline exhaust		Diesel exhaust		
	Euro2	Euro3, 4 and 5	Euro2	Euro3	Euro4 and 5
Alkanes	31% ($\pm 5\%$)	29% ($\pm 5\%$)	24% ($\pm 6\%$)	22% ($\pm 6\%$)	21% ($\pm 6\%$)
Alkenes	17% ($\pm 3\%$)	16% ($\pm 3\%$)	16% ($\pm 12\%$)	15% ($\pm 12\%$)	15% ($\pm 12\%$)
Alkynes	3% ($\pm 1\%$)	3% ($\pm 1\%$)	2% ($\pm 3\%$)	2% ($\pm 3\%$)	2% ($\pm 3\%$)
Aldehydes	3% ($\pm 2\%$)	3% ($\pm 2\%$)	29% ($\pm 12\%$)	28% ($\pm 12\%$)	26% ($\pm 12\%$)
Ketones	1% ($\pm 0\%$)	1% ($\pm 0\%$)	4% ($\pm 1\%$)	4% ($\pm 1\%$)	3% ($\pm 1\%$)
Aromatics	44% ($\pm 9\%$)	42% ($\pm 9\%$)	18% ($\pm 3\%$)	17% ($\pm 3\%$)	17% ($\pm 3\%$)
PAH	$\leq 1\%$ (nd)	6% (nd)	7% (nd)	12% (nd)	16% (nd)

default continental compartment of USEtox cannot necessarily be applied to specific regions of the world. For instance, Middle East environmental characteristics, where crude oil is extracted, are different from Europe: e.g. less urban zone, less rain falls, higher temperatures and less fresh water reserves. As a consequence, the dispersion of a substance among the various compartments of the environment (atmosphere, soil and water) varies. Therefore, it has been decided to create new regional compartments, according to where crude oil and hard coal are extracted. Figure 2 shows on a world map the seven zones (RU, Russia; RME, Middle East; RNAF, North Africa; RCAF, Central Africa; RSAF, South Africa; AU, Australia; RCAM, Central America) that have been defined while Table 5 describes their characteristics (only the characteristics differing from

the default continental scale are shown). These characteristics are calculated using the GLOBACK model (Sleeswijk 2011), which provides all the data needed to calculate landscape data for new regions in USEtox, except sea areas which are arbitrarily calculated using a geographical information system (GIS).

2.5 Atmospheric emissions: urban and rural compartments

To calculate the repartition between urban and rural emissions in the WTT stage, it is necessary to know the geographical coordinates of all emitting sources and to have a map of urban and rural areas. These data are then incorporated into a GIS tool. Data of urban and rural areas are obtained from the Global Rural–urban Mapping Project

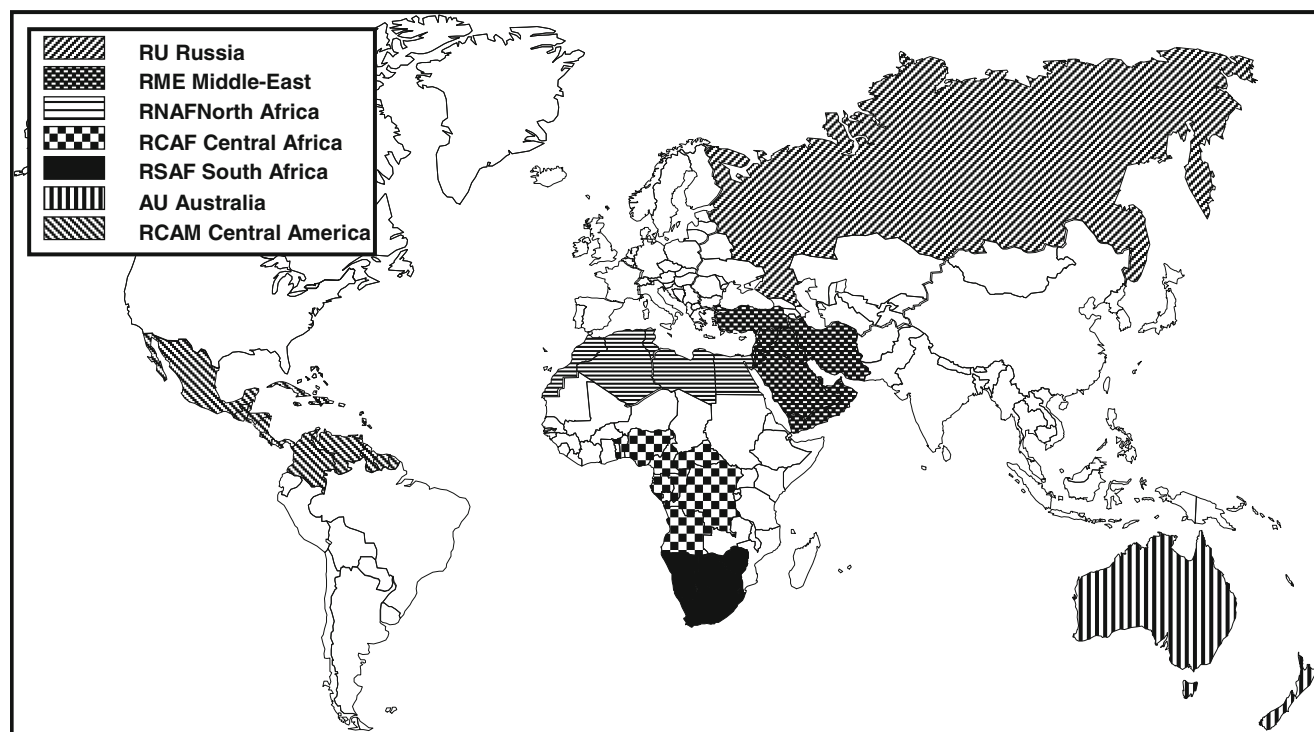
**Fig. 2** USEtox new calculated regions (Mercator projection)

Table 5 Main characteristics of the seven region compartments

Zone	RU	RME	RNAF	RCAF	RSAP	AU	RCAM
Area (km ²)	17,075,400	7,008,264	5,752,891	6,364,053	3,083,998	7,686,850	4,932,750
Population (habitat)	141,800,000	347,293,844	161,238,500	268,272,273	68,538,392	22,475,500	256,876,203
Temperature (°C)	4	24	19	25	19	13	19
Rainfall (mm an ⁻¹)	460	188	99	1,378	453	534	1,686
Agric fraction (%)	7	8	4	8	6	3	8
Sea (km ²)	17,000,000	3,000,000	2,700,000	3,300,000	1,100,000	10,000,000	19,700,000

(GRUMP) of the SocioEconomic Data and Application Centre.² The resolution of this project is 30" (corresponding to 925 m for longitude), allowing accurate results. Figure 3 illustrates the GIS developed: refineries (circles), combustion power plants (triangles) and hard coal mines (stars), whose coordinates are obtained from the European Pollutant Release and Transfer Register,³ are placed on the GRUMP map of Europe with urban (grey) and rural (white) areas.

For emissions linked to transport, the shares of rural and urban emissions is calculated according to the repartition of rural and urban zones of the regions crossed. Emissions outside the EU27 are located according to different sources. The coordinates of oil-extraction places are obtained from Elvidge et al. (2009) while hard coal mines come from <http://www.mbendi.com>. Emissions linked to crude oil and coal transport to the EU27 are located according to the regions crossed between the EU27 and the production zones. The repartition of filling stations is considered identical to the repartition of the population between rural and urban in the EU27. For the emissions of all processes that are not directly linked to the production of the fuels, it is not feasible to calculate the exact ratio between rural and urban areas using a GIS. As these impacts are less relevant than the emissions directly associated with the system studied, it was decided that this repartition would be equal to the general repartition of the continent when the processes are located.

3 Results

3.1 Significant impacts

USEtox contains characterization factors for atmospheric, aquatic and soil emissions. In the three pathways studied, there is no significant emission into the soil. Table 6 shows the share of the impacts, among freshwater, rural air, seawater and urban air and between organics (NMVOCs) and inorganics (heavy metals). At this stage, no rural/urban

distinction is applied to the sources of emissions, which means that one half of atmospheric emissions is assigned to the rural compartment, the other half being assigned to the urban one. As a result, the sum of emissions of all compartments is equal to 100%. Not surprisingly, emissions into the air compartment account for the majority of the impacts. This is an expected conclusion because the studied pathways mainly deal with the combustion of fossil fuels (crude oil, gasoline, diesel and hard coal). The only exception comes from emissions of inorganics in freshwater by hard coal electricity which are caused by leakages during the mining of coal. Therefore, the detailed results for aquatic ecotoxicity and human health are only given for atmospheric emissions in the next paragraphs.

3.2 Urban/rural differentiation

For the WTT stage, the share of rural and urban emissions is calculated according to the GIS tool developed, as described in the "Materials and methods". Table 7 shows the share of urban emissions, according to the WTT stage. Rural share is equal to the remaining share.

For the TTW stage, two options are retained. We consider that emissions and fuel consumption between rural and urban driving are the same. However, urban and rural emissions are distinguished by entirely assigning the emissions into the urban compartment for the first case and into the rural compartment for the second case. Obviously, this distinction is only valid for diesel- and gasoline-powered cars because the battery-powered car does not emit any pollutant during its use.

3.3 Aquatic ecotoxicity

Figure 4 shows the WTW results for aquatic ecotoxicity caused by atmospheric emissions. For inorganics, there is no significant difference in the results between rural and urban emissions (less than 1%). Nevertheless, a difference (+85% for gasoline and +88% for diesel) between rural and urban organics is observed. For both organics and inorganics, ICE cars are more impacting than the electric vehicle because of the exhaust pipes of the cars (the WTT

² <http://sedac.ciesin.columbia.edu/gpw/index.jsp>

³ <http://prtr.ec.europa.eu/>

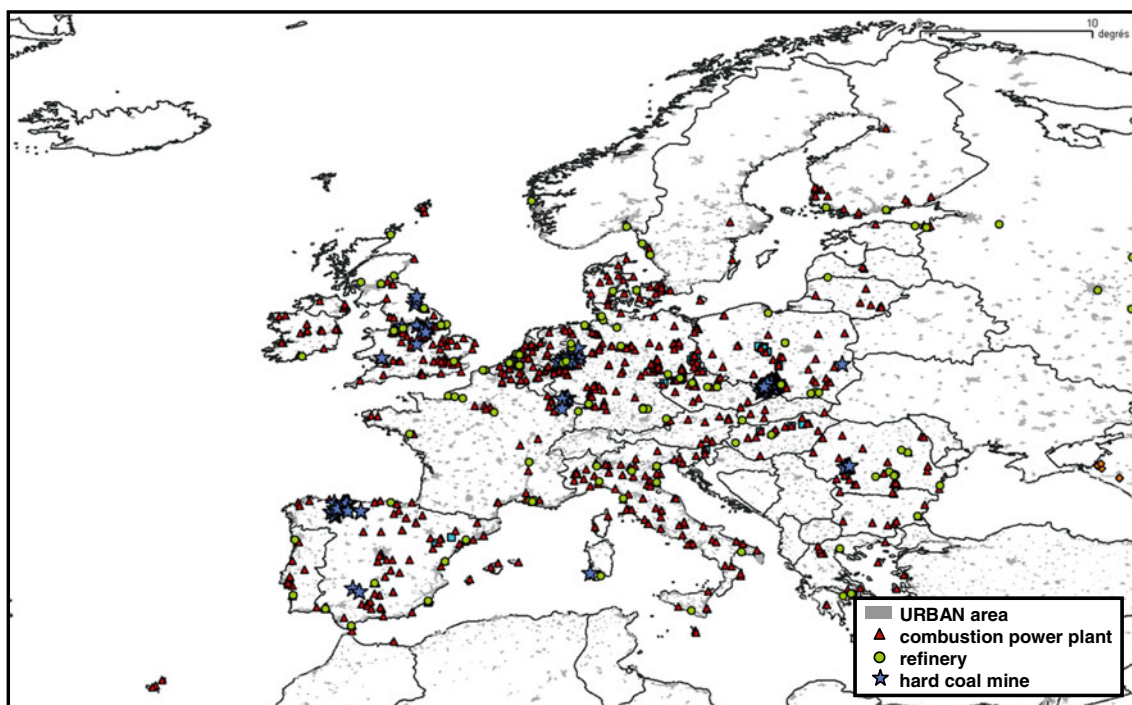


Fig. 3 Refineries, combustion power plants and hard coal mines on GRUMP map (extended Lambert projection)

part being insignificant). For organics, this impact is mainly due to PAH and aldehyde emissions. The Monte Carlo uncertainty analysis shows that these differences are significant. Yet, because the uncertainty analysis does not take into account PAHs, these conclusions are uncertain. Ecotoxicity caused by organics is lower than ecotoxicity from inorganics. For inorganics, gasoline and diesel impacts come equally from the WTT and the TTW stages. Heavy metals responsible for ecotoxicity are copper (64% for gasoline, 67% for diesel and 17% for hard coal), vanadium (3% for gasoline, 2% for diesel and 41% for hard coal), zinc (17% for gasoline and diesel and 11% for hard coal), nickel (11% for gasoline, 9% for diesel and 16% for hard

coal) and, to a lesser extent, chromium, arsenic and selenium. Uncertainties associated with these emissions are high but gasoline and diesel are nonetheless significantly more impacting than coal electricity.

3.4 Human health

Figures 5 and 6 show the WTW results for human health impacts. They are separated between organics and inorganics and between cancer (see Fig. 5) and non-cancer (see Fig. 6) effects.

For inorganics, variations between rural and urban emissions are not significant (+5%). Inorganics that are

Table 6 WTW impacts according to the receiving compartment and between organics and inorganics

Compartment of emission	Human health			Ecotoxicity		
	Gasoline (%)	Diesel (%)	Coal (%)	Gasoline (%)	Diesel (%)	Coal (%)
Freshwater, inorganics	5	3	3	13	12	38
Freshwater, organics	≤1	≤1	≤1	≤1	≤1	≤1
Seawater, inorganics	≤1	≤1	≤1	≤1	≤1	≤1
Seawater, organics	≤1	≤1	≤1	≤1	≤1	≤1
Rural air, inorganics	38	19	49	41	36	31
Rural air, organics	1	2	≤1	2	6	≤1
Urban air, inorganics	36	18	48	40	35	31
Urban air, organics	20	57	≤1	3	11	≤1
All compartments	100	100	100	100	100	100

Table 7 Urban shares of WTT stages

WTT stage	Urban share
Crude oil extraction	0–81% (depending on the region)
Crude oil and hard coal transportation	0–77% (depending on the region)
Refineries	86%
Diesel and gasoline distribution	76%
Hard coal extraction	0–86% (depending on the region)
Hard coal power plants	77%

responsible for the impacts are different from those responsible for ecotoxicity issues: zinc (67% for ICE cars and 19% for hard coal), mercury (21% for ICE cars and 65% for hard coal) and, to a lesser extent, arsenic, chromium, lead and cadmium. As a consequence, hard coal electricity is more impacting, contrary to ecotoxicity results. However, uncertainties associated with inorganics are high for ICE fuels (they are inferior for hard coal because the uncertainties associated with mercury emissions are lower). Coal mines and power plants are the main sources of inorganic pollution while for gasoline and diesel, human health impacts mainly come from oil refining and car use and are equivalent because they use the same resource (crude oil) and the heavy metal content of gasoline and diesel fuel is no different.

Organics results show that the main sources of pollutants are the emissions associated with diesel- and gasoline-powered cars (WTT impacts are low). This assertion is valid for both cancer and non-cancer issues. NMVOCs causing these impacts are from the aldehyde group (acrolein, methacrolein and formaldehyde), which have cancer and non-cancer effects. The difference between gasoline and diesel is lower for cancer effects because gasoline also emits higher amounts of aromatic compounds (benzene) which are highly carcinogenic. Nevertheless, non-cancer effects are about ten times higher than cancer

effect and, as a result, offset them in the overall human health impact. The difference between urban and rural emissions is high for gasoline and diesel fuel (more than ten times) and diesel has the highest impacts, because of its aldehyde emissions (see “Materials and methods”). This conclusion is confirmed by the Monte Carlo uncertainty analysis, which shows that these differences are significant. Hard coal impacts are negligible because, according to our data, no aldehydes are produced during the combustion of coal.

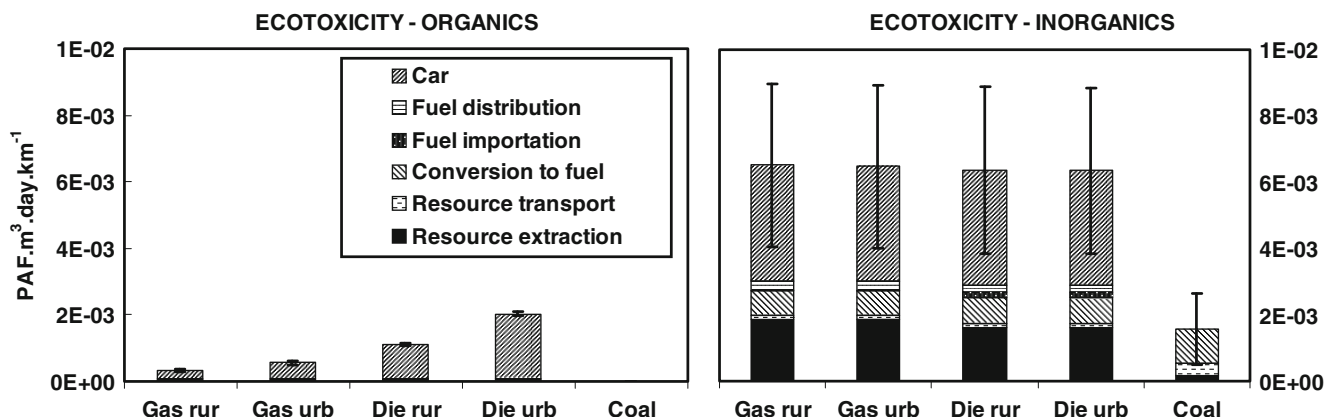
3.5 Comparison with other indicators

Table 8 shows the final results of USEtox in comparison with three other methods (as presented in the introduction paragraph). Because the units differ among the methods, it is necessary to normalize the results. Gasoline rural WTW results are thus considered as being equal to 1 for each methodology and the other fuel impacts are calculated accordingly. This allows to rank the three fuels, depending on the methodology considered.

Results among the three methods do not lead to the same conclusions. For instance, for aquatic ecotoxicity, CML 2001 ranks hard coal power as the less impacting pathway, followed by diesel fuel and gasoline. Using EDIP 1997 leads to the conclusion that all pathways are almost equal while IMPACT 2002+ ranks hard coal power as the most impacting fuel. For ecotoxicity, USEtox results are close to CML2001 while for human health, the ranking among the three fuels completely differs. Thus, Table 8 emphasizes the need for a consensual methodology.

3.6 Compartment differentiation

The new continental compartments as defined in the “Materials and methods” do not lead to major changes in the overall WTW results. This is due to the fact that crude oil extraction and hard coal mining have low impacts

**Fig. 4** WTW ecotoxicity results

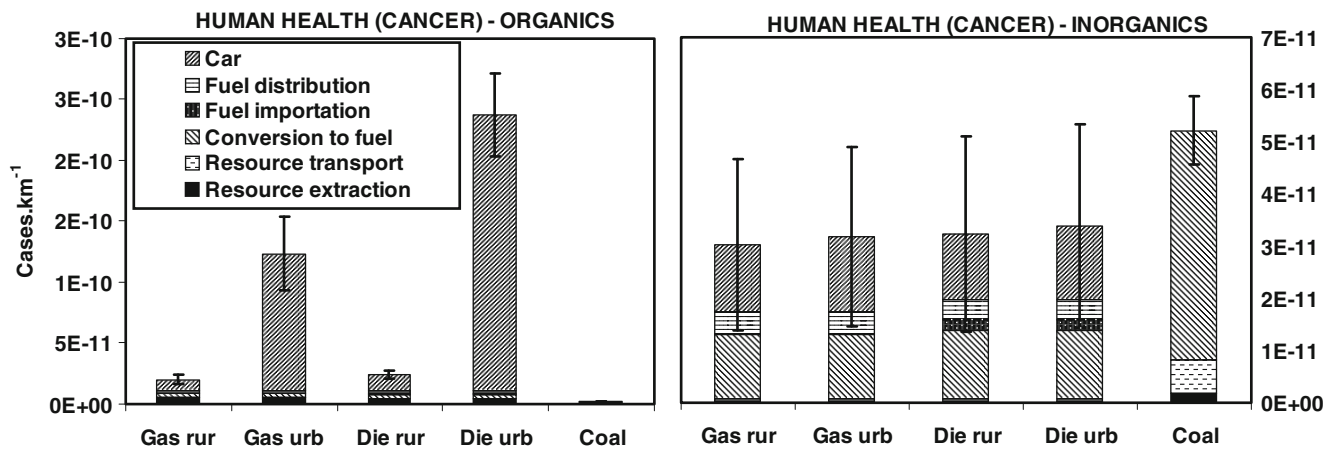


Fig. 5 WTW human health (cancer) impact results

compared with other stages (such as, for instance, oil refining or car exhausts). Even so, changing the compartments does change the characterization factors. In order to see these differences, Table 9 focuses on the crude oil-extraction stage for the gasoline pathway. The first column shows the default values while the second one shows the values with region-depending characterization factors. The new characterization factors increase the results for both aquatic ecotoxicity and human health. Arbitrarily defining the sea area had no effect on the impact values because seawater emissions are negligible.

4 Discussion

4.1 Organics

USEtox human health impacts are mainly linked to the emissions of aldehydes and, to a much lesser extent, aromatics. Aromatics are only impacting for cancer issues, which are by ten times inferior than non-cancer impacts,

and still not as impacting as aldehydes (which are both carcinogenic and non-carcinogenic). Aldehydes have a low lifetime in the environment, compared with PAH. This means that, for car exhausts, the effects assessed by USEtox are especially caused by inhalation. This assertion is confirmed by the clear distinction between rural and urban impacts, the latter being for diesel fuel more than ten times higher than rural impact. When comparing the three fuels, the differences between those used in ICEs (gasoline and diesel) and hard coal electricity are of several orders of magnitude, the latter being clearly less impacting. ICEs used in a rural context also have low impact values but these values increase by ten times when assessed in an urban context, ranking diesel fuel as the most impacting pathway. Gasoline, which is commonly thought to be harmful because of benzene and other aromatic compounds, has a lower impact value because USEtox cancer effects are lower than non-cancer effects.

Aquatic ecotoxicity is caused by aldehyde and PAH emissions and coal electricity has the lowest impacts. Compared with human health, differences between diesel

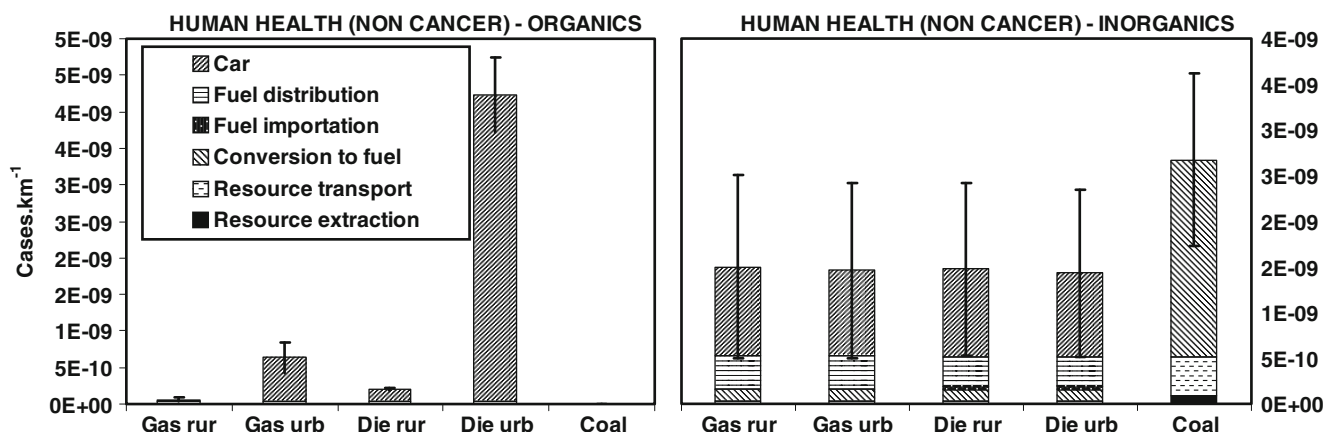


Fig. 6 WTW human health (non-cancer) impact results

Table 8 Comparison of USEtox WTW results with other indicators

	Gasoline		Diesel fuel		Hard coal power
	Rural	Urban	Rural	Urban	
Aquatic ecotoxicity					
CML 2001	1.0	1.0	3.6	3.6	0.3
EDIP 1997	1.0	1.0	0.8	0.8	0.8
IMPACT 2002+	1.0	1.0	1.2	1.2	2.7
USEtox, organics	1.0	1.7	3.3	6.0	≤0.1
USEtox, inorganics	1.0	1.0	1.0	1.0	0.3
Human health					
CML 2001	1.0	1.0	0.4	0.4	0.3
EDIP 1997	1.0	1.0	1.2	1.2	0.1
IMPACT 2002+	1.0	1.0	0.3	0.3	0.2
USEtox, organics	1.0	20.8	5.7	115.4	≤0.1
USEtox, inorganics	1.0	1.0	1.0	1.0	1.8

and gasoline decrease, because PAH are emitted by both pathways. The difference between rural and urban values (less than twice) is also far less important than for the human health indicator because aquatic toxicity is obviously not caused by inhalation but by ingestion through the trophic chain in the aquatic milieu. Considering that uncertainties tend to increase when mechanisms are being more complex (inhalation is simpler than transport through different compartments of the environment and accumulation throughout the trophic chain), this means that ecotoxicity values are less robust than human health impacts. Furthermore, PAH uncertainties being unknown, no conclusions can be drawn for this indicator.

4.2 Inorganics

As described in the USEtox model, inorganics characterization factors are all considered as interim values. It is indeed emphasized that the mechanisms determining the toxicity of heavy metals are more complex than for organic compounds. The differences between urban air and rural air values are very low, although for human health the number of targets in the air compartment is much more important in urban than in rural area. This means that the toxicity of heavy metals in USEtox is mainly caused by the ingestion routes through the food chain rather than by direct inhalation.

Dispersion in the environment and accumulation through the food chain are complex mechanisms to assess. As a consequence, uncertainties are great with inorganics impacts since they are almost entirely dependent of these mechanisms. Furthermore, uncertainties increase because of the high persistence of heavy metals in ecosystems. Contrary to organic compounds, heavy metals are not biodegradable and the only mechanism that can prevent them from being available to the ecosystem is sedimentation. Once absorbed, they are bioaccumulated through the trophic chain. However, the fraction between absorbed and sedimented heavy metals cannot be easily determined as it involves various physical and chemical mechanisms. The near-zero biodegradation rate used in USEtox is artificially increasing the effect of ingestion compared with inhalation (for human health issues) and overrating inorganics compared with organics. Moreover, uncertainties associated with heavy metals emissions in the atmosphere are high because of the variability of heavy metals contained in burned fuels.

Thus, we do not recommend, when conducting an automotive fuel LCA, to sum the impacts of inorganics and organics as the high characterization factors could offset the impact of organic compounds while uncertainties make the interpretation too complex. If required, inorganic factors should be used with care.

Table 9 Characterization factors differentiation according to the region compartment for crude oil extraction for gasoline

Impact (km ⁻¹)	Default value	Region-depending value
Aquatic ecotoxicity, inorganics	6.4×10^{-4} PAF m ⁻³ day ⁻¹	1.9×10^{-3} PAF m ⁻³ day ⁻¹
Aquatic ecotoxicity, organics	3.9×10^{-5} PAF m ⁻³ day ⁻¹	5.6×10^{-4} PAF m ⁻³ day ⁻¹
Human health, inorganics	1.5×10^{-12} cases	3.0×10^{-12} cases
Human health, organics	3.0×10^{-12} cases	5.2×10^{-12} cases

5 Conclusions and further researches

5.1 USEtox relevance as an impact indicator for automotive fuels

USEtox is an effective tool to assess the toxic and ecotoxic impacts of fuel pathways, especially for NMVOC impacts. Almost all NMVOCs emitted by the three pathways are included in the organics database and the missing substances did not affect the results. The difficulty comes from the fact that NMVOC emissions are usually only expressed as NMVOCs with no regards of the species emitted. Though it can be sufficient to assess photochemical ozone formation, this has no relevance for calculating USEtox impacts. Therefore, for NMVOC emissions linked with combustion, a special care must be taken when using USEtox. In this article, the speciation of NMVOCs is as comprehensive as possible, allowing the assessment of the species that are mainly responsible for the impact of the fuels. However, when comparing alternative fuels with conventional fossil fuels, the LCA practitioner might not have all the information on NMVOC composition. Table 10 shows, for a given fuel pathway, the minimum level of details required for a WTW comparison with fossil fuels.

Heavy metals emissions can be more easily calculated since the content of most fuels is given by the EMEP/EEA guidebook. Contrary to organics, the difficulties are linked to the interpretation of the results. Because of the high uncertainties associated with the characterization factors and with the emissions values, results for inorganics cannot be summed with organics. Further researches are therefore needed for this impact to be used by the automotive industry.

It must be kept in mind that USEtox, for emissions associated with fuel combustion (especially the combustion of fuels by car engines), is only addressing one of the several human health and ecotoxicity issues of automotive fuels. NMVOCs also have a strong effect on photochemical ozone formation, which can afterwards have sanitary effects, as described in the “Materials and methods”. NO_x as well play a role in this impact while also creating secondary particulate matter (along with PM and SO₂) and acidification (with SO₂). USEtox human health values must be used in addition with methodologies such as CML2001,

IMPACT2002+ or ReCiPe2008. For ecotoxicity, USEtox is only considering the aquatic compartment. However, as in the TRACI methodology, assessment of air ecotoxicity should also be included in USEtox since considering only aquatic ecotoxicity might lead to an underestimation of the ecotoxic impacts of fuels, mainly linked to emissions into the atmosphere.

5.2 Urban and rural distinction

It is obvious that the impacts of cars on human health are higher in cities than in rural places. However, USEtox is the first methodologies that allow to further study this assertion. It allows to compare, on a consensual basis, the impacts of (for instance) diesel cars used in urban context with electric vehicles. It provides a useful methodology to assess the impact of zero-emission vehicles (electricity or hydrogen powered cars) on human health or hybrid vehicles than can use ICE in road-driving while being powered by electricity in city driving. This is especially useful as these technologies are now emerging in daily life.

5.3 Further researches

Further researches should explore four directions. First, as mentioned above, USEtox is an indicator that can be used to assess human health issues associated with automotive fuels, especially because of the rural/urban distinction of atmospheric emissions. However, as explained in the USEtox relevance section, it is not sufficient to assess all human health issues associated with cars. Thus, a similar distinction for impacts such as ReCiPe2008 particulate matter formation potential would be necessary in order to tend towards a more comprehensive human health impact.

Secondly, in this article, we show the possibility of distinguishing urban from rural emissions thanks to a GIS. This GIS should be extended to other systems, such as the whole car, in order to assess the feasibility in general LCA of such an approach. If it is possible to know the origin of the main atmospheric emissions sources, the urban/rural distinction might be a step towards more accurate LCA.

Thirdly, this rural/urban distinction should also be extended to the emission inventory because cars emit different amounts of pollutants, whether they are driven in

Table 10 Level of details required for using USEtox in a fuel LCA

Accuracy	Level of analysis
No NMVOC speciation	USEtox cannot be used
Aldehyde speciation	Rough USEtox human health assessment
Aldehyde+HAP speciation	Human health and aquatic ecotoxicity assessment
Aldehyde+HAP+aromatic speciation	Cancer effect calculation
Complete speciation	All USEtox effects caused by organics

rural or in urban contexts. Associating the variations in emissions with the appropriate compartment would increase the accuracy of fuel LCA.

Finally, USEtox should be tested on other fuel systems, in order to know if the maximum level of details required in Table 9 can also be obtained. Assessing fuels such as biofuels would also allow assessing the significance of the car use compared with the pesticide use during the agriculture step. Moreover, it would show if aquatic and soil emissions can be compared with atmospheric emissions.

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